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DEVELOPMENT AND APPLICATIONS OF TUNABLE, NARROW BAND LASERS AND STIMULATED RAMAN SCATTERING DEVICES FOR ATMOSPHERIC LIDAR

by

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Technical Note BN-1150

June 1993

INSTITUTE FOR PHYSICAL SCIENCE AND TECHNOLOGY

DEVELOPMENT AND APPLICATIONS OF TUNABLE, NARROW BAND LASERS AND STIMULATED RAMAN SCATTERING DEVICES FOR ATMOSPHERIC LIDAR

Final Report on NASA Cooperative Agreement NCC1-25

by

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June 1993

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DEVELOPMENT AND APPLICATIONS OF TUNABLE, NARROW BAND LASERS AND STIMULATED RAMAN SCATTERING DEVICES FOR ATMOSPHERIC LIDAR

ABSTRACT

We summarize the results of several research projects carried out under NASA Cooperative Agreement NCC1-25. The main thrust of the program has been the study of stimulated Raman processes for application to atmospheric lidar measurements. This has involved the development of suitably narrow, tunable lasers, the detailed study of stimulated Raman scattering, and the use of the Raman-shifted light for new measurements of molecular line strengths and line widths. The principal spectral region explored in this work has been the visible and near-IR wavelengths between 500 nm and 1.5 μm. Recent alexandrite ring laser experiments are reported, involving diode injection-locking, Raman shifting, and frequency-doubling, that succeeded in producing tunable light at 577 and 937 nm with linewidths in the range 80-160 Mhz.

ACKNOWLEDGEMENTS

The Principal Investigator wishes to acknowledge the contributions of many others to this work: Drs. John Theon and Ramesh Kakar of NASA Headquarters for their funding support, Dr. Edward V. Browell of NASA-Langley for his many years of support and encouragement both as a colleague and a grant monitor, Leo Cotnoir and Drs. Upendra Singh, Zhiping Chu, and Rita Mahon of the University of Maryland for their extensive technical and scientific contributions, Alfred J. Fay and Prof. James Yorke of the University of Maryland for moral and financial support of the program, Scott Higdon and Dr. Benoist Grossmann of NASA-Langley for their participation in a key experiment, and Stefan Schmitz and Prof. U.von Zahn (University of Bonn) and Drs. Donald Heller, John Walling, and Georgia Fisanick of Light Age, Inc. for generous collegial and facilities contributions in the final phase of the grant research.

1. INTRODUCTION

The research summarized in this report is a follow-on to several years of joint work with the NASA-Langley Research Center on ground based and airborne lidar (DIAL) for profiling water vapor in the atmosphere. Most of the research was carried out in the Atmospheric Lidar Observatory at Maryland. The final part of the work was conducted in the laboratories of Light Age, Inc. in Somerset, NJ.

Mainly we have undertaken to develop light sources that could be used for water vapor DIAL measurements in the H_2O bands at wavelengths of 900 nm and longer, in order to lay a foundation for water vapor lidar in the stratosphere and the polar troposphere where the H_2O abundance is far less than in the troposphere at mid- and tropical latitudes. In very dry air, one can obtain good DIAL performance only by operating at the longer wavelengths where the increased H_2O line strengths compensate for the lower molecular concentrations, so as to maintain reasonable values of the absorption optical depth.

Two studies of tunable, narrow band lasers and Raman shifting employed a dye laser which, when shifted in hydrogen and deuterium, provided narrow band ($\Delta\nu \leq 0.03$ cm^{-1}) collision-dominated Raman radiation out to wavelengths of order 980 nm.

As part of this program we have also taken advantage of opportunities to add to our understanding of stimulated Raman (SRS) processes, to apply some of the generated wavelengths to atmospheric observations and molecular spectroscopy, and to explore the high spectral resolution possibilities offered by a relatively new type of laser operating in conjunction with Raman shifting and harmonic generation.

At 532 nm, we carried out an extensive study of the SRS efficiencies in H_2, D_2, and CH_4 for the various Stokes and anti-Stokes lines, in order to establish a basis for possible multiwavelength aerosol lidar systems.

At 1.06 μm, efficient first Stokes generation of eyesafe 1.54 μm radiation in CH_4 was made possible for the first time by means of backward-wave "self-seeding" of the SRS process.

Quantitative spectroscopic measurements (line strength and line width) were made on H_2O lines in the range 960-980 nm, using the narrow band, tunable emission from the Maryland dye laser Raman-shifted in hydrogen.

Journal publications on the above are briefly summarized in Section 2. Dr. Chu's doctoral dissertation is outlined in Section 3. The most recent work involving the alexandrite laser, which as yet has been published only as extended conference abstracts, is described in greater detail in Section 4 and Appendices A & B.

2. PUBLISHED ARTICLES

(A) *Applied Optics* **26**, 1617-1621 (1987):

Raman-shifted dye laser for water vapor DIAL measurements

B. E. Grossmann, U. N. Singh, N. S. Higdon, L. J. Cotnoir, T. D. Wilkerson, and E. V. Browell

For improved DIAL measurements of water vapor in the upper troposphere or lower stratosphere, we have generated narrowband (~0.03-cm^{-1}) laser radiation at 720- and 940-nm wavelengths by stimulated Raman scattering (SRS) using the narrow linewidth (~0.02-cm^{-1}) output of a Nd:YAG-pumped dye laser. For a hydrogen pressure of 350 psi, the first Stokes conversion efficiencies to 940 nm were 20% and 35% when using a conventional and waveguide Raman cell, respectively. We measured the linewidth of the first Stokes line at high cell pressures and inferred collisional broadening coefficients that agree well with those previously measured in spontaneous Raman scattering.

(B) *Optics Communications* **75**, 173-178 (1990):

A SELF-SEEDED SRS SYSTEM FOR THE GENERATION OF 1.54 μm EYE-SAFE RADIATION

Z. CHU, U.N. SINGH and T.D. WILKERSON
University of Maryland, Institute for Physical Science and Technology, College Park, MD 20742-2431, USA

Received 30 August 1989; revised manuscript received 11 November 1989

A light source is described for the efficient generation of 1.54 μm for eye-safe aerosol lidar operation. The system is based upon a Nd:YAG laser at 1.06 μm which is then Raman-shifted in methane to produce light at the first Stokes wavelength of 1.54 μm. First Stokes light generated in the backward direction was retroreflected back into the Raman cell for amplification in the tail of the 10-ns pump beam. The energy conversion efficiency and the spatial beam quality of the amplified first Stokes were found to be adversely affected when operated at higher repetition rates due to a thermal gradient produced in the generation region. A Stokes energy of 25 mJ was obtained for a pumping energy of 140 mJ at a repetition rate of 10 Hz. The beam divergence of the amplified Stokes radiation was measured to be less than 1 mrad. The optimized results demonstrate the applicability of this radiation for eye-safe lidar measurements.

(C) *Applied Optics* **29**, 1730-1735 (1990)

Optimization of a Raman shifted dye laser system for DIAL applications

Upendra N. Singh, Zhiping Chu, Rita Mahon, and Thomas D. Wilkerson

We describe an efficient Raman shifted dye laser system that generates tunable radiation at 765 and 940 nm with a bandwidth of 0.03 cm^{-1}. Operating a Raman cell at hydrogen pressure below 14 atm, we recorded optimum first Stokes energy conversions of 45% and of 37% at 765 and 940 nm, respectively. Optical depth measurements made at the centers of twenty-five absorption lines in the *P* branch of the oxygen *A* band imply a high spectral purity for both the laser and the Raman shifted radiation, and thus indicate the feasibility of using the stimulated Raman scattered radiation for differential absorption lidar (DIAL) measurements.

(D) *Applied Optics* **30**, 4350-4357 (1990):

Multiple Stokes wavelength generation in H_2, D_2, and CH_4 for lidar aerosol measurements

Zhiping Chu, Upendra N. Singh, and Thomas D. Wilkerson

We report experimental results of multiple Stokes generation of a frequency-doubled Nd:YAG laser in H_2, D_2, and CH_4 in a focusing geometry. The energies at four Stokes orders were measured as functions of pump energy and gas pressure. The characteristics of the Stokes radiation generated in these gases are compared for optical production of multiple wavelengths. The competition between Raman components is analyzed in terms of cascade Raman scattering and four-wave mixing. The results indicate the possibility of using these generation processes for atmospheric aerosol measurements by means of multiwavelength lidar systems. Also this study distinguishes between the gases, as regards the tendency to produce several wavelengths (H_2, D_2) versus the preference to produce mainly first Stokes radiation (CH_4).

Key words: Stimulated Raman scattering, multiwavelength generation, aerosol lidar application, lidar, Raman shifting in molecular gases, pressure dependence of Raman conversion.

(E) *Applied Optics* **32**, 992-998 (1993)

Water-vapor absorption line measurements in the 940-nm band by using a Raman-shifted dye laser

Zhiping Chu, Thomas D. Wilkerson, and Upendra N. Singh

We report water-vapor absorption line measurements that are made by using the first Stokes radiation (930–982 nm) with HWHM 0.015 cm^{-1} generated by a narrow-linewidth, tunable dye laser. Forty-five absorption line strengths are measured with an uncertainty of 6% and among them are fourteen strong lines that are compared with previous measurements for the assessment of spectral purity of the light source. Thirty air-broadened linewidths are measured with 8% uncertainty at ambient atmospheric pressure with an average of 0.101 cm^{-1}. The lines are selected for the purpose of temperature-sensitive or temperature-insensitive lidar measurements. Results for these line strengths and linewidths are corrected for broadband radiation and finite laser linewidth (0.015 cm^{-1} HWHM) broadening effects and compared with the high-resolution transmission molecular absorption.
Key words: Water vapor, absorption lines, line strength, line width, Raman-shifted laser, water-vapor spectroscopy.

3. PhD DISSERTATION OF DR. ZHIPING CHU

Dr. Chu was awarded the Allen Prize of the Optical Society of America for her graduate student research on optical remote sensing, which appears in the papers above and in the dissertation, which has the following **Abstract**:

RESEARCH ON STIMULATED RAMAN SCATTERING WITH APPLICATIONS TO ATMOSPHERIC LIDAR

Research has been conducted on stimulated Raman scattering (SRS) to extend conventional lasers into the infrared where lidar systems can make important contributions to observations of the atmosphere. An efficient "Raman shifted" dye laser system was used to generate tunable and narrow band radiation

at 760 and 940 nm for differential absorption lidar applications. The requisite tunability and spectral purity of the output is derived from the dye laser input by controlling the Raman cell at pressure below 14 atm. The converted radiation is optimized for different pump focusing geometries. Energy conversion efficiencies of 45% and 37% at 765 and 940 nm, respectively, were obtained. Optical depth measurements and calculations were made at the centers of 25 lines in the P branch of the oxygen A-band in air. The data and theoretical calculations agree, indicating a high spectral purity of the light source. High resolution parameters of water vapor at 940 nm were obtained using this narrow linewidth Raman-shifted dye laser in conjunction with a multi-pass optical absorption call. Optical strengths and Lorentz widths were deduced from the data using a Voigt line profile to numerically correct for finite laser linewidth. Some lines are compared with prior measurements by Giver *et al* that used a wholly different method. Some lines , which were not covered in Giver's experiments, were compared with Hitran database. The simultaneous generation of several Stokes orders was investigated in H_2, D_2, and CH_4, for the purpose of multiple wavelength lidar. The study was focused on the redistribution of the pump energy into the different SRS components. Optimal experimental conditions were investigated and calculated. Eye-safe radiation at 1.54 μm was generated for lidar applications, by Raman shifting Nd:YAG laser light (1.06 μm) in methane. To increase conversion efficiency, a novel self-seeding oscillator and amplifier system was designed and used. Backward first Stokes radiation was separated and used as seed by being refocused into the Raman cell and amplified by the rest of the pump pulse. A maximum conversion efficiency of 18% was obtained.

4. TUNABLE, NARROW BAND ALEXANDRITE LASER DEVELOPMENT FOR LIDAR

The author was invited by Dr. Donald Heller of Light Age, Inc. to work as a guest scientist in that firm's laboratory in Bound Brook, NJ, to investigate new alexandrite laser techniques for obtaining laser output that is spectrally narrow enough for lidar remote sensing. This work took place at the end of the grant period, and was elected over the prior concept of experiments at Maryland using dye lasers or Raman-shifted Nd:YAG. The opportunity became available to use combinations of diode injection-locking of the alexandrite, Raman shifting (using the proprietary Light Age cells), and harmonic generation. We adopt the common term "Raman shifting" to refer to *stimulated* (as opposed to spontaneous) Raman scattering.

Our goal was to obtain tunable, narrow band radiation in the H_2O absorption bands near 720, 940, and 1140 nm, with line widths of order 0.01 cm^{-1} (300 MHz). For reasons of scheduling, not all these wavelengths could be covered at this time, and some of the work was done at other wavelengths to illustrate the general capabilities of these combined, alexandrite-based systems. One factor was the availability of diodes at certain frequencies. Another factor was the timing of simultaneous work at Light Age by Mr. Stefan Schmitz, a student of Prof. U.von Zahn of Bonn University. It took some months to work out schedules so that Mr. Schmitz and the author could work together; moreover, the contractual work for Bonn had to concentrate on methods leading to narrow band lidar operation in the atomic sodium D-line (Na D_2) near 589 nm. So a significant portion of our joint efforts addressed this wavelength region, on the good assumption that the developed techniques will readily transfer to any wavelength lying within the operating range of the alexandrite laser, nominally 720-820 nm.

This short R&D program has been very successful, and several of the results are summarized in this report. We have described the work at the OSA meeting in Salt Lake City (March '93) and the CLEO meeting in Baltimore (May '93) ; the extended abstracts for these meetings are reprinted here in Appendices **A** and **B**. The first part of our studies employed the linear alexandrite oscillator configuration typical of Light Age's 101 PAL system ; a major advance was then made by going to a new ring alexandrite laser, which has given the most exciting results of very narrow band emission. Line widths of order 10^{-3} cm^{-1} (~ 30 MHz) have been achieved. Measurements involving Raman frequency conversion employed the Light Age proprietary Raman cells, 101 PAL/RC, filled with H_2 or D_2 at pressures of order 15 - 20 atmospheres.

(A) Linear Alexandrite Oscillator

A schematic of this arrangement is shown in **Figure 1**. The wavelengths indicated apply to relatively long-wave operation of the alexandrite near 790 nm, so as to obtain H_2-Raman cell output (first Stokes) at 1178 nm which, when frequency-doubled, converts to 589 nm. The diagnostics we used initially for the output radiation were a 1" Tec-Optics Fabry-Perot system from the University of Maryland together with a small CCD camera, a VCR, and (for capturing individual frames of the VCR tapes) either a TV monitor that

LIGHT AGE, INC.

INJECTION SEEDED ALEXANDRITE LASER FOR PRODUCING 589 NM FOR NA LIDAR

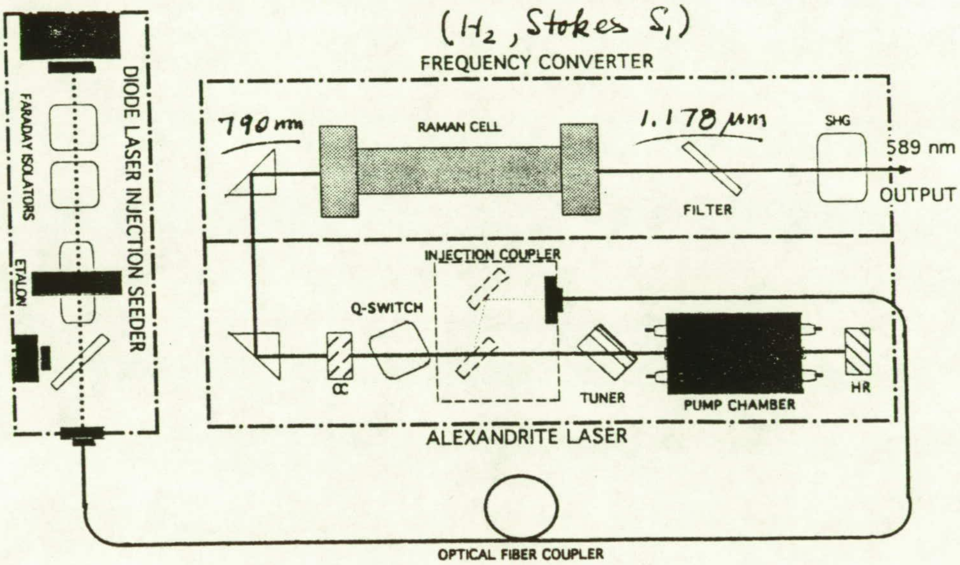


FIGURE 1

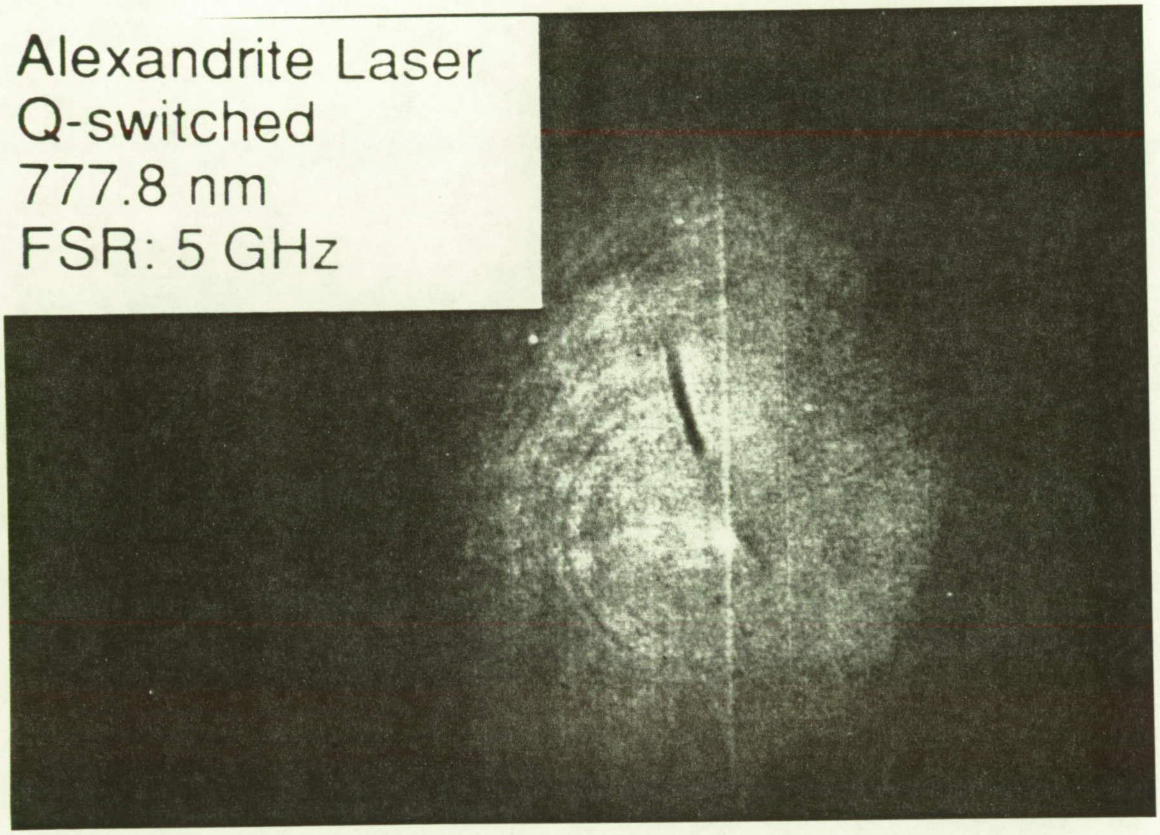


FIGURE 2

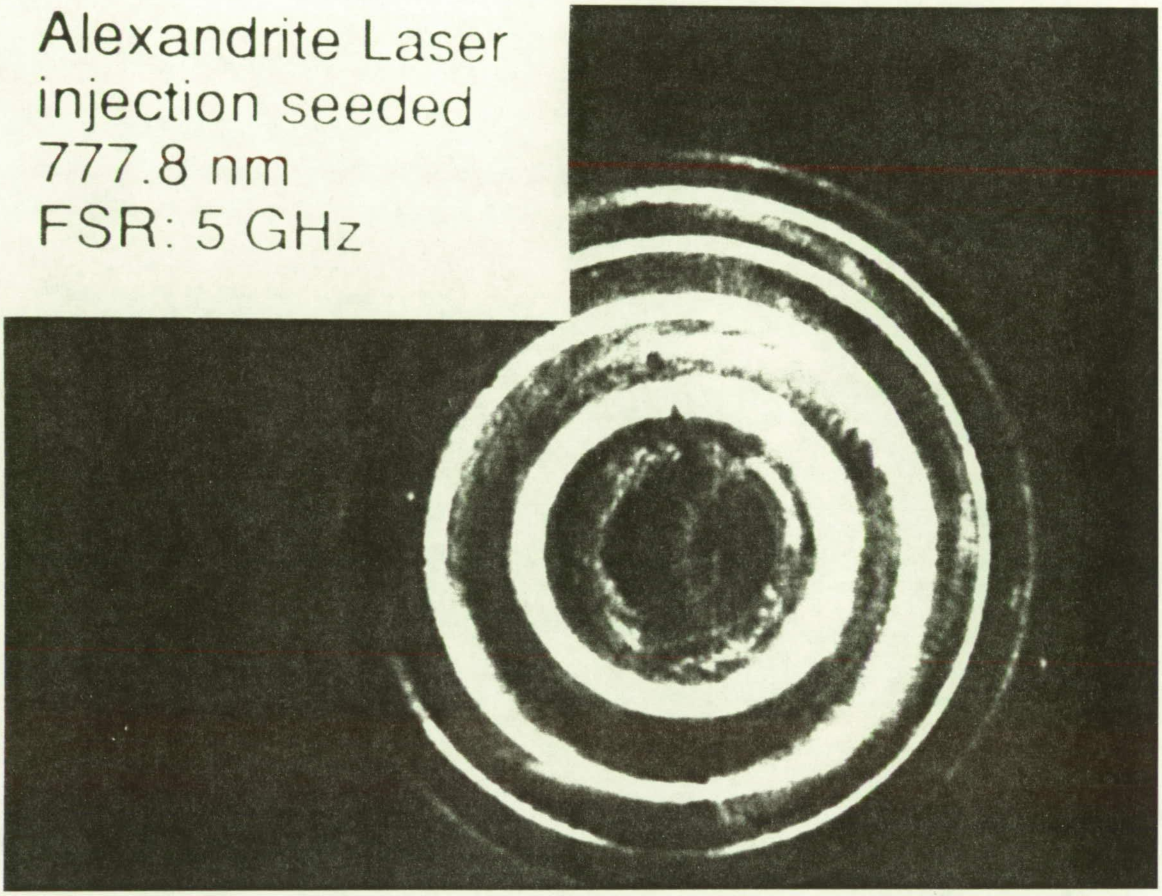


FIGURE 3

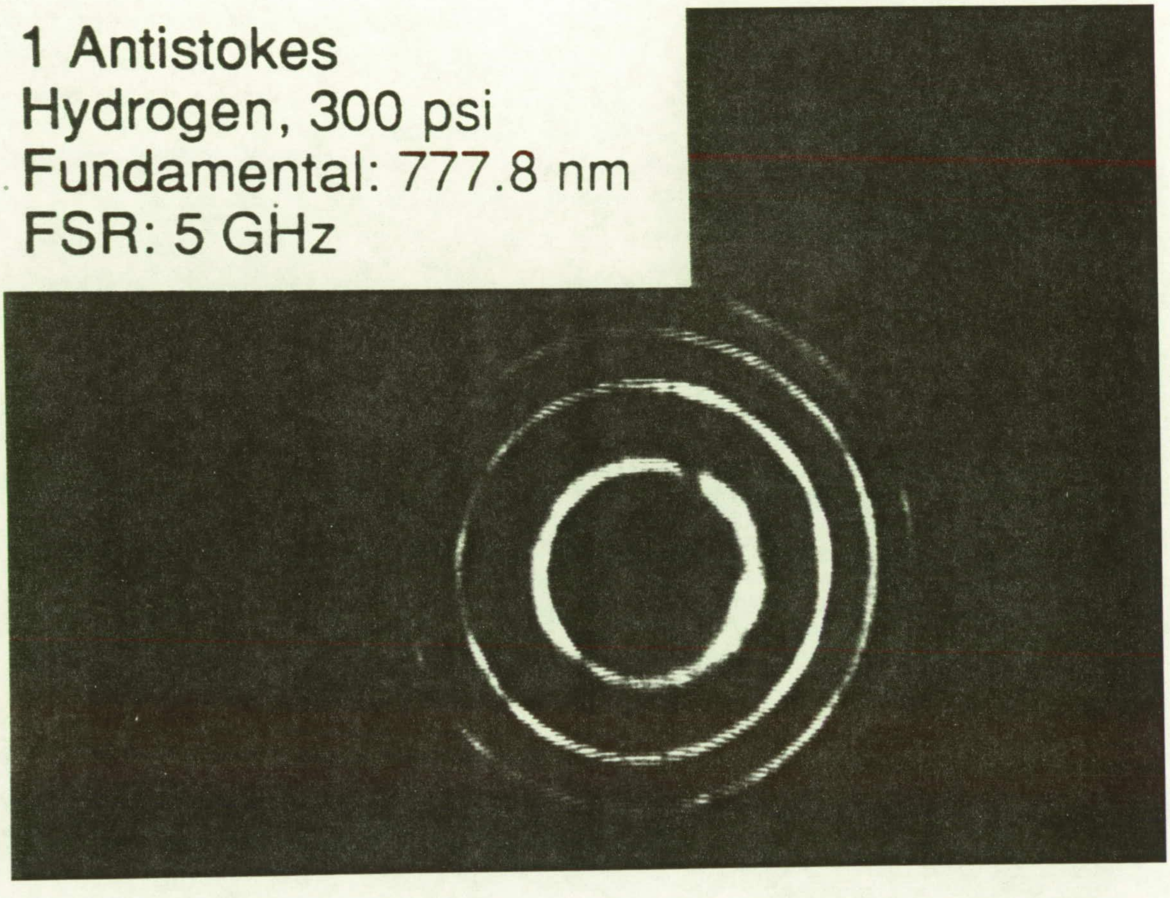


FIGURE 4

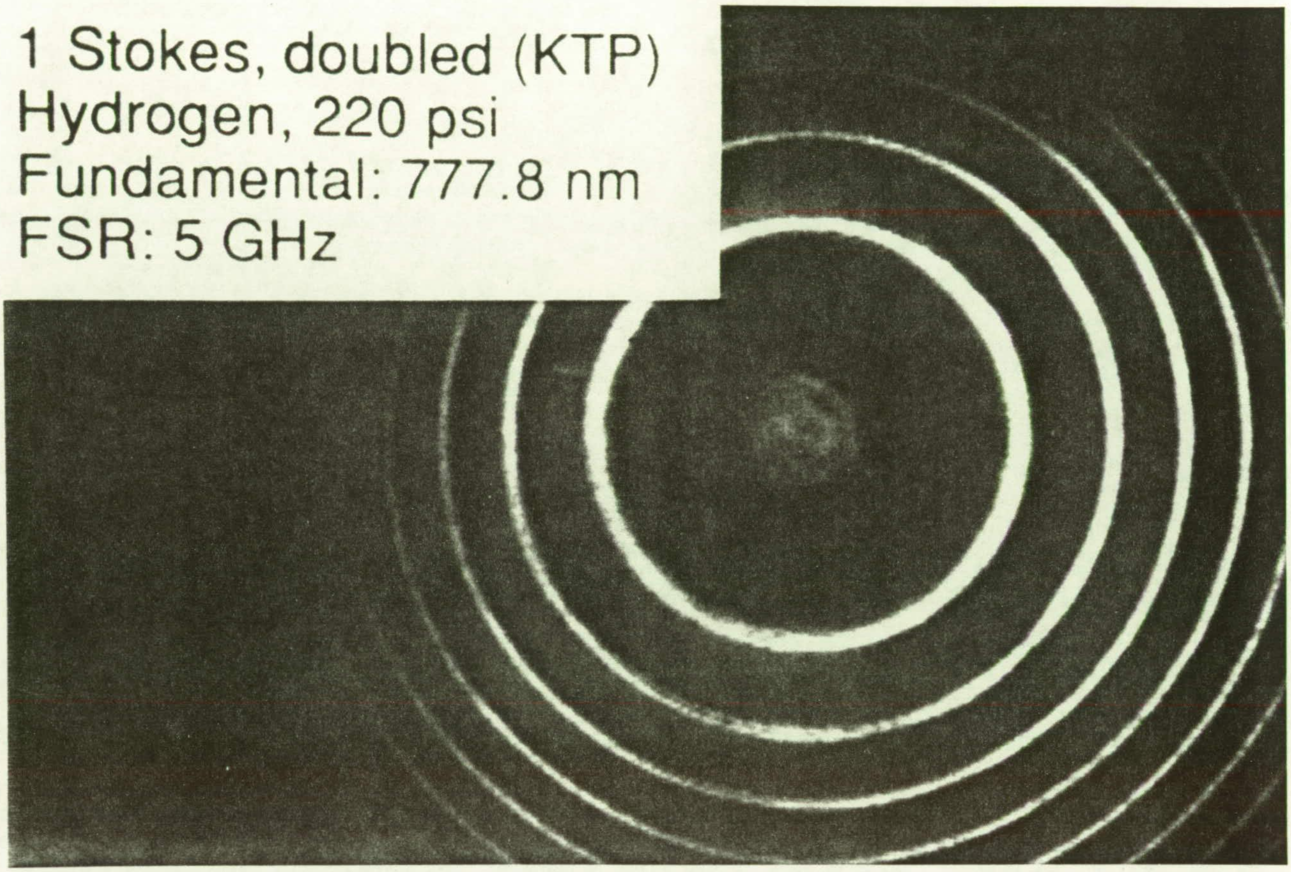
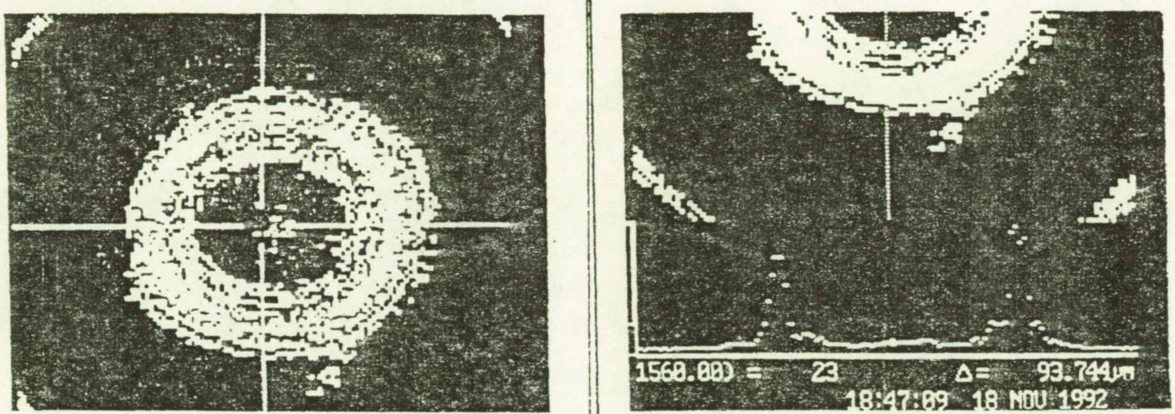

1 Stokes, doubled (KTP)
Hydrogen, 220 psi
Fundamental: 777.8 nm
FSR: 5 GHz

FIGURE 5

SINGLE MODE OUTPUT FROM ALEXANDRITE LINEAR RESONATOR 780.14



ALEXANDRITE LINEWIDTH <80 MHZ FWHM

MEASURED INSTRUMENTAL LINEWIDTH <40 MHZ

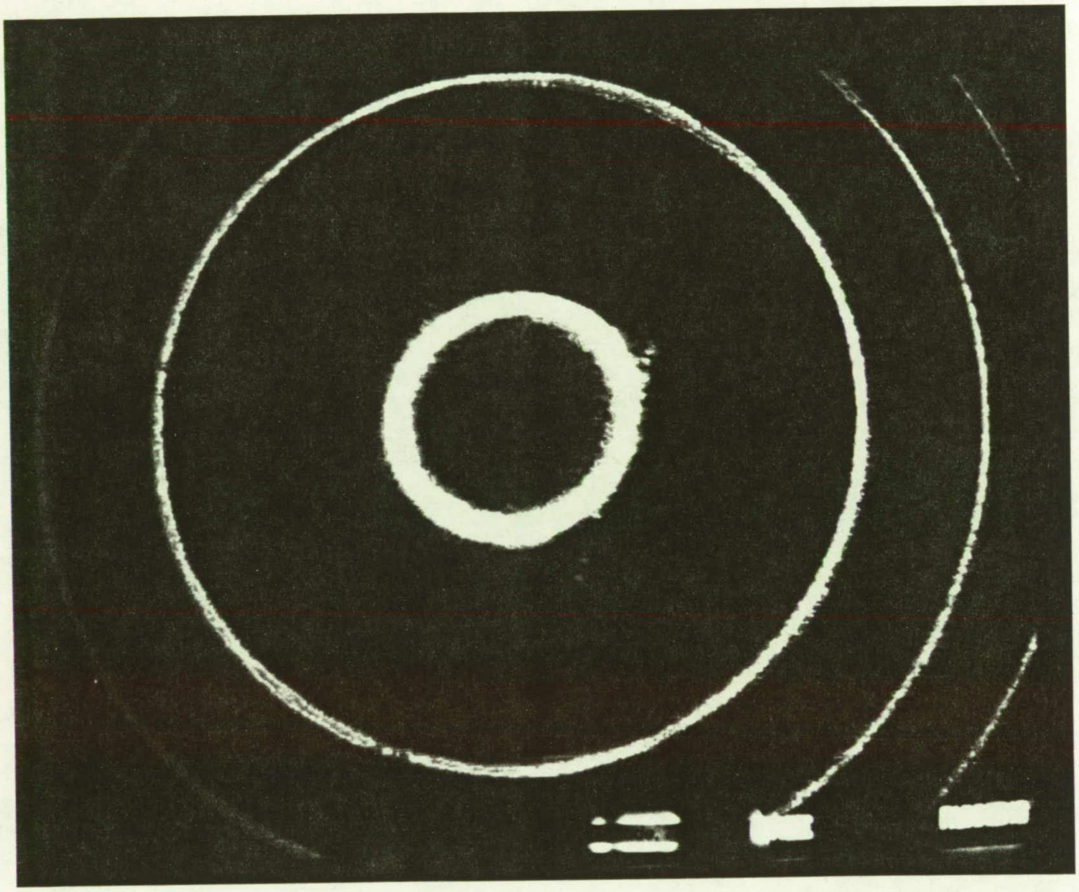
5 GHZ FREE SPECTRAL RANGE

20 MHZ INSTRUMENTALRESOLUTION LIMIT

FIGURE 6

SINGLE MODE OUTPUT FROM LASER DIODE

SPECTRA DIODE LABS #SDL5411-G1 100MW
780.14 NM



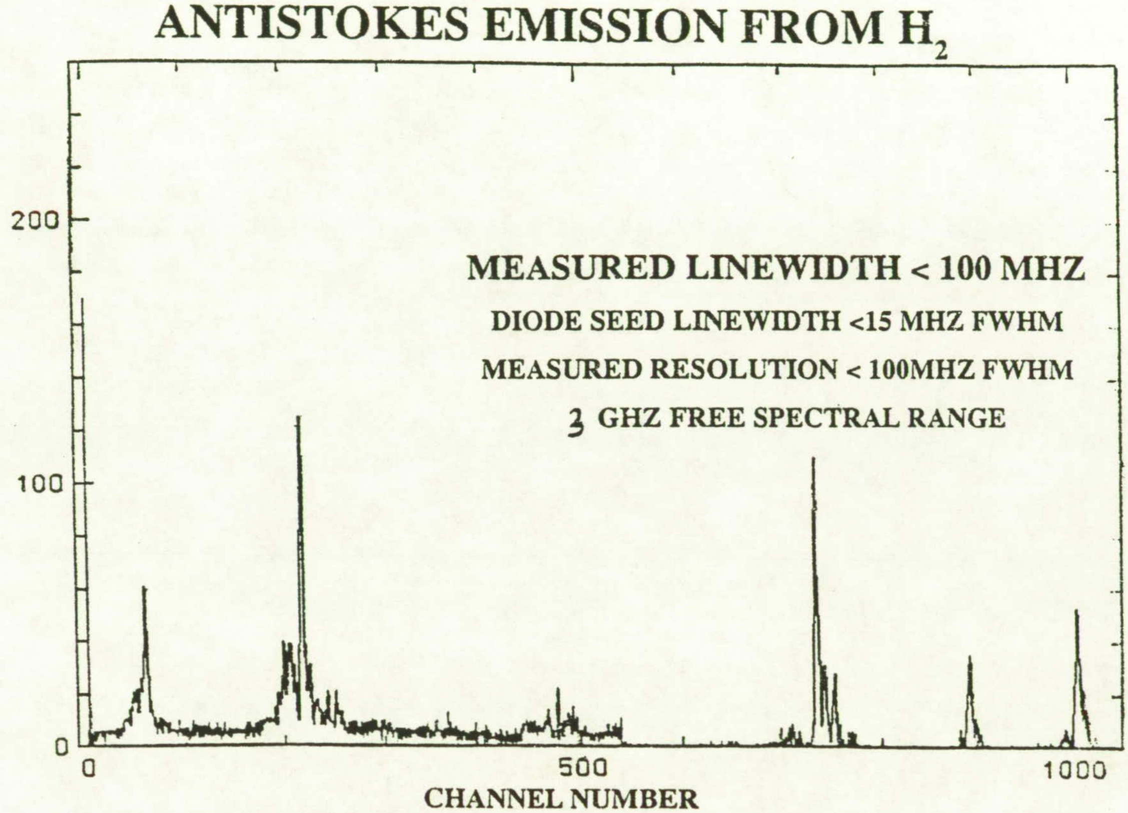
DIODE LINEWIDTH <40 MHZ FWHM

DIODE LINEWIDTH < 15 MHZ FWHM(PUBLISHED SPECIFICATION)

5 GHZ FREE SPECTRAL RANGE

20 MHZ INSTRUMENTALRESOLUTION LIMIT

FIGURE 7



LINES ARE INSTRUMENTALLY LIMITED TO 100 MHZ

FIGURE 8

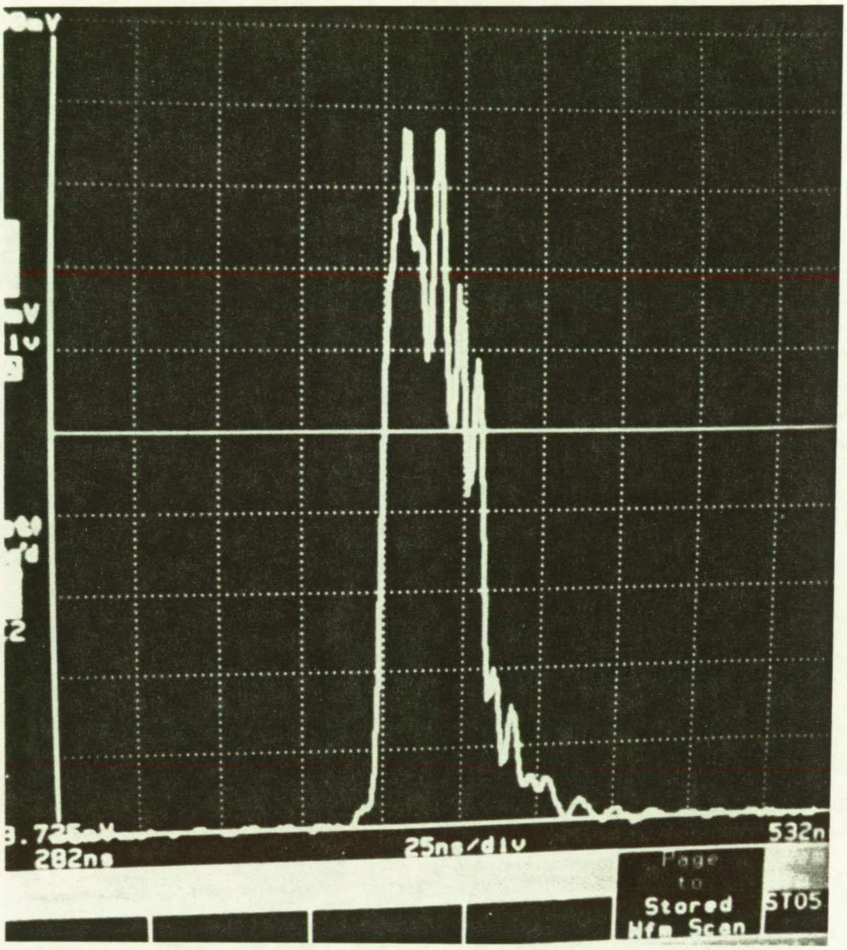


FIGURE 9

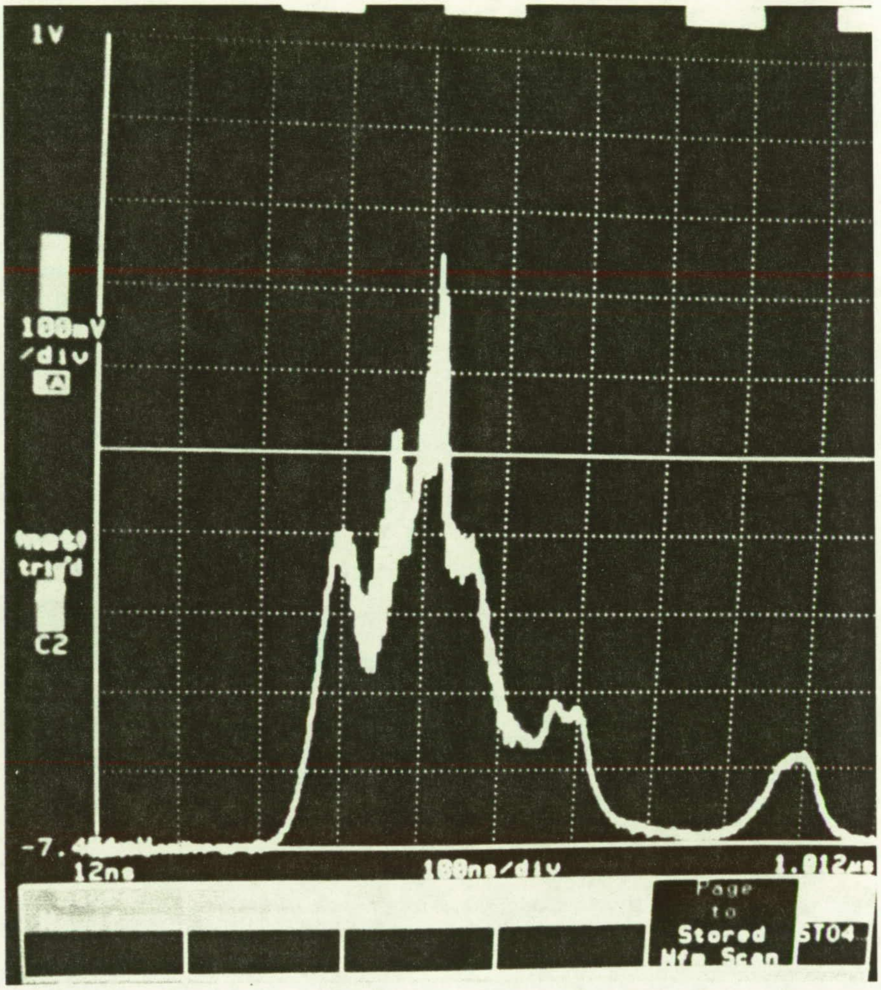

1V
100mV
/div
trig'd
C2
12ns
100ns/div
Page
to
Stored
Wfm Scan
ST04

FIGURE 10

LIGHT AGE, INC.

INJECTION SEEDED RING LASER

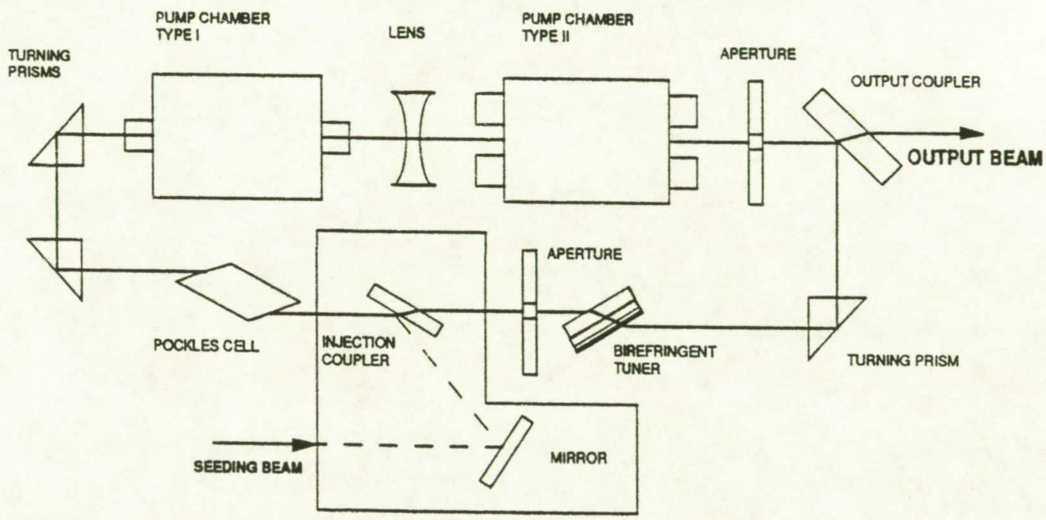


FIGURE 11

was photographed or a Tektronix thermal printer. Later we used a Spiricon beam profiling imager for both the spatial pattern of the beam and the Fabry-Perot fringe patterns. **Figures 2 - 5**, respectively, show for λ = 777.8 nm the fringe patterns for Q-switched alexandrite (no injection seeding), the injection-seeded output, Raman-shifted radiation (H_2 anti-Stokes) at 588 nm, and frequency-doubled (KTP) Raman-shifted light (H_2 first Stokes) at 575 nm. These data were obtained with the video system, and the frequency resolution (on linewidth) is at best about 350-380 MHz. While the video system and the small interferometer were convenient for initial work, it was clear that their performance was not good enough for definitive work at the level of 100 MHz and below.

When the Spiricon imager was employed with a Burleigh 2" Fabry-Perot system, much better instrumental resolution could be obtained. **Figure 6** demonstrates that, when the alexandrite linear oscillator is running in a single spectral mode, its linewidth is less than 80 MHz ; the corresponding diode injection spectrum is shown in **Figure 7**, where we can conclude that the diode linewidth is surely below 40 MHz, consistent with the vendor's specification of this diode. **Figure 8** shows a Burleigh wavemeter spectrum of the corresponding H_2-Raman (anti-Stokes) output, which has a linewidth $\langle$ 100 MHz.

Typically the output of the linear alexandrite oscillator varied between 1 and 4 longitudinal modes, partly because we made no attempt to "trim" the diode frequency to the oscillator mode frequencies or *vice versa*. We could just barely detect the presence of the separate modes with the first imaging system, as their spacing was about 200 MHz.

While the performance of the linear oscillator was variable, we nonetheless demonstrated that the Raman linewidths, being in the range 100-350 MHz for H_2, are about 1/10 - 1/4 the values for the spontaneous Raman linewidths at the cell pressures employed. Thus substantial gain-narrowing was confirmed, even with improvements still to be made in the alexandrite oscillator. As many as 5 orders of anti-Stokes beams and 2 orders of Stokes beams were observed. Pump depletion was as great as 50% in some cases, and Stokes conversion greater than 30% could be obtained ; anti-stokes conversion in excess of 5% was observed. **Figure 9** demonstrates the onset of Raman emission, which is quite sudden relative to the time-dependence of the alexandrite input to the Raman cell.

One difficulty with the linear oscillator is that the standing wave pattern leads to hole-burning. A symptom of this is shown in **Figure 10** where, in spite of the diode seeding, the laser's light output becomes quite variable. We therefore changed to a ring laser configuration to obtain greater temporal and spectral stability.

(B) Alexandrite Ring Laser

The two-chamber ring laser configuration is shown schematically in **Figure 11**, with injection seeding. The expected improvement with this design arises from the circulating radiation wave pattern, as opposed to the standing wave property of the linear cavity which leads to hole-burning.

Figure 12 shows the Fabry-Perot fringe pattern for the single spectral mode output of the diode-injection seeded alexandrite ring laser near 780 nm. The linewidth is less than 40 MHz, which is our limit on linewidth resolution in this case. It should be noted that here we are not operating at the optimum wavelength for alexandrite output; nonetheless, very good performance of the laser is achieved.

The effect of seeding on the beam profile (TEM_{00}) and energy of the alexandrite ring laser is shown in **Figures 13** and **14.** Relative to the beam pattern of the unseeded laser (**Fig. 13**), we see about twice the beam energy with the pattern pulled in much more tightly in **Fig. 14**. Also, effective seeding decreases the cavity buildup time by about 50-70 nsec.

The presence of two spectral modes in the alexandrite ring laser is indicated in **Figure 15**. Finally, when only one mode is propagating the light emission from the ring follows the relatively smooth trace shown in **Figure 16**.

The most favorable ring operation we have encountered so far showed the energy ratio between the desired propagation direction and the counter-propagating radiation to be about 11. **Figure 17** is interesting because it shows the relative timing of the intensity buildup in the two directions, with the counter-propagating one rising later due to scattering of the main beam into the reverse direction. (Neither the intensity scales nor the absolute buildup times shown here are meaningful.)

The best linewidth results obtained to date with Raman-shifting of the seeded alexandrite ring laser are as follows:

(1) $\Delta\nu$ = 80 MHz (2.7×10^{-3} cm^{-1}) for λ_{alex} = 780 nm,
Stokes shifted (H_2) and frequency doubled to 577 nm

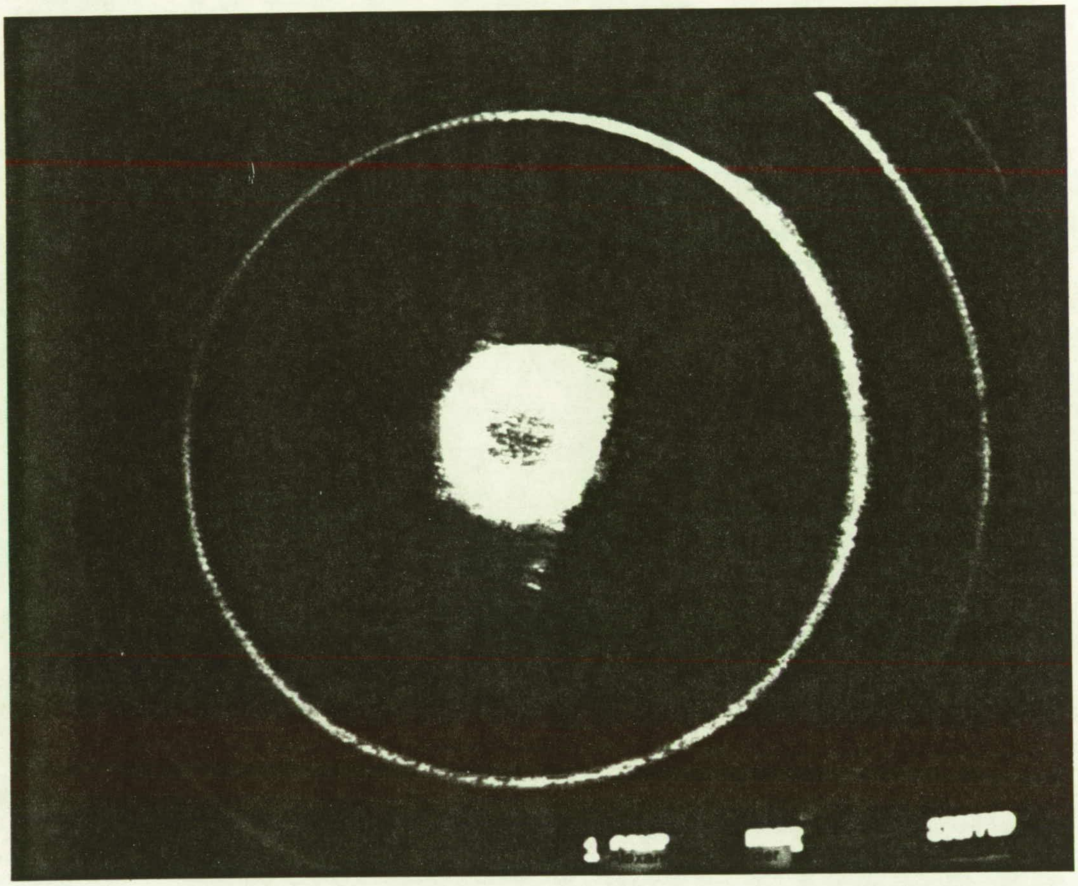
(2) $\Delta\nu$ = 160 MHz (5.4×10^{-3} cm^{-1}) for λ_{alex} = 732 nm,
Stokes shifted (D_2) to 937 nm

Thus it has been demonstrated that one can obtain very narrow band, tunable light suitable for lidar studies of sodium (mesosphere) and water vapor (troposphere, stratosphere), using the alexandrite ring laser together with Raman shifting and harmonic generation. The linewidths are far less than the pressure-broadened widths one calculates for spontaneous Raman scattering, and are thus suited to the narrow line resonance and absorption processes employed in atmospheric lidar observations.

(C) Discussion, Conclusions and Implications

It is conceivable that transform-limited linewidths can be obtained using systems of the type described above. As pointed out by Don Heller, the foundation for that possibility can be seen in the sum of the reports by other researchers. First, David King (NIST, private communication to Light Age) has injection-locked an alexandrite laser with the

SINGLE MODE OUTPUT FROM ALEXANDRITE RING RESONATOR 780.14NM



ALEXANDRITE LINEWIDTH <40 MHZ FWHM

MEASURED INSTRUMENTAL LIMIT < 40MHZ FWHM

5 GHZ FREE SPECTRAL RANGE

20 MHZ INSTRUMENTALRESOLUTION LIMIT

FIGURE 12

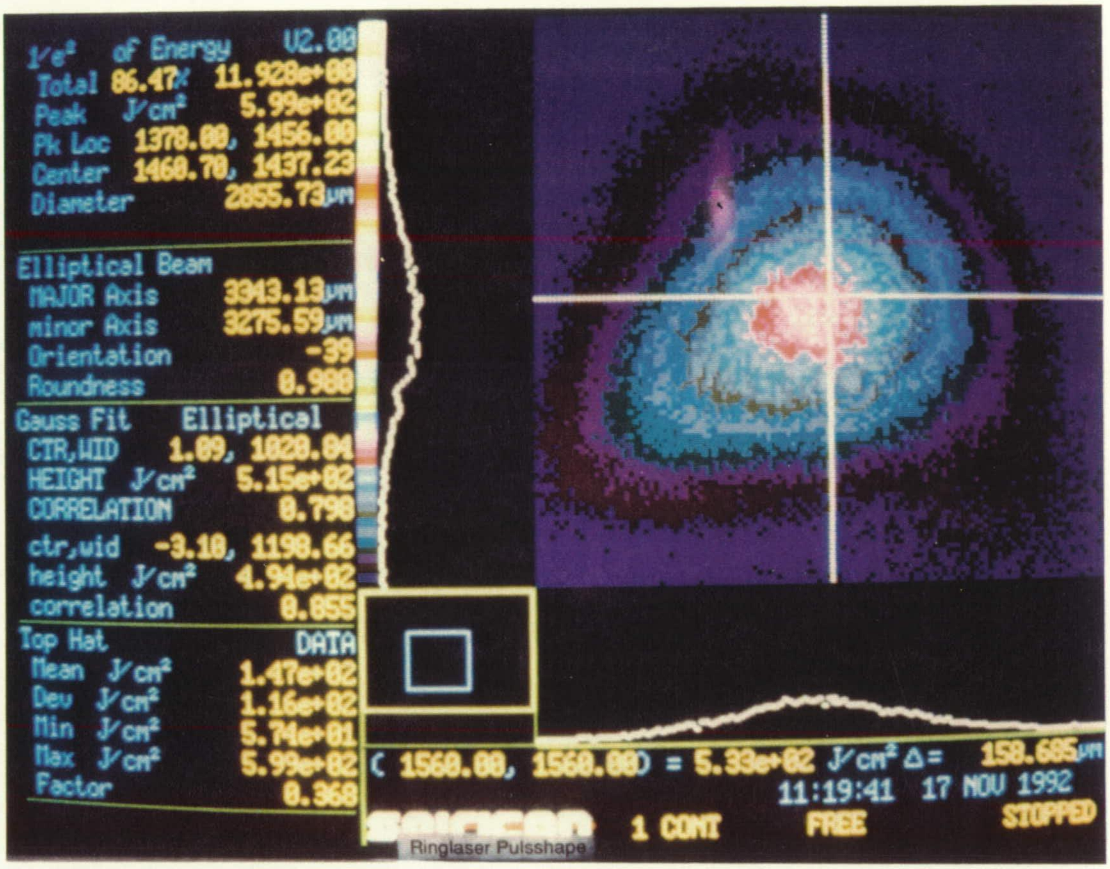

1/e² of Energy V2.00
Total 86.47% 11.928e+00
Peak J/cm² 5.99e+02
Pk Loc 1378.00, 1456.00
Center 1460.70, 1437.23
Diameter 2855.73µm
Elliptical Beam
MAJOR Axis 3343.13µm
minor Axis 3275.59µm
Orientation -39
Roundness 0.980
Gauss Fit Elliptical
CTR,WID 1.09, 1020.04
HEIGHT J/cm² 5.15e+02
CORRELATION 0.798
ctr,wid -3.10, 1198.66
height J/cm² 4.94e+02
correlation 0.855
Top Hat DATA
Mean J/cm² 1.47e+02
Dev J/cm² 1.16e+02
Min J/cm² 5.74e+01
Max J/cm² 5.99e+02
Factor 0.368
(1560.00, 1560.00) = 5.33e+02 J/cm² Δ= 158.685µm
11:19:41 17 NOV 1992
1 CONT
FREE
STOPPED
Ringlaser Pulsshape

FIGURE 13

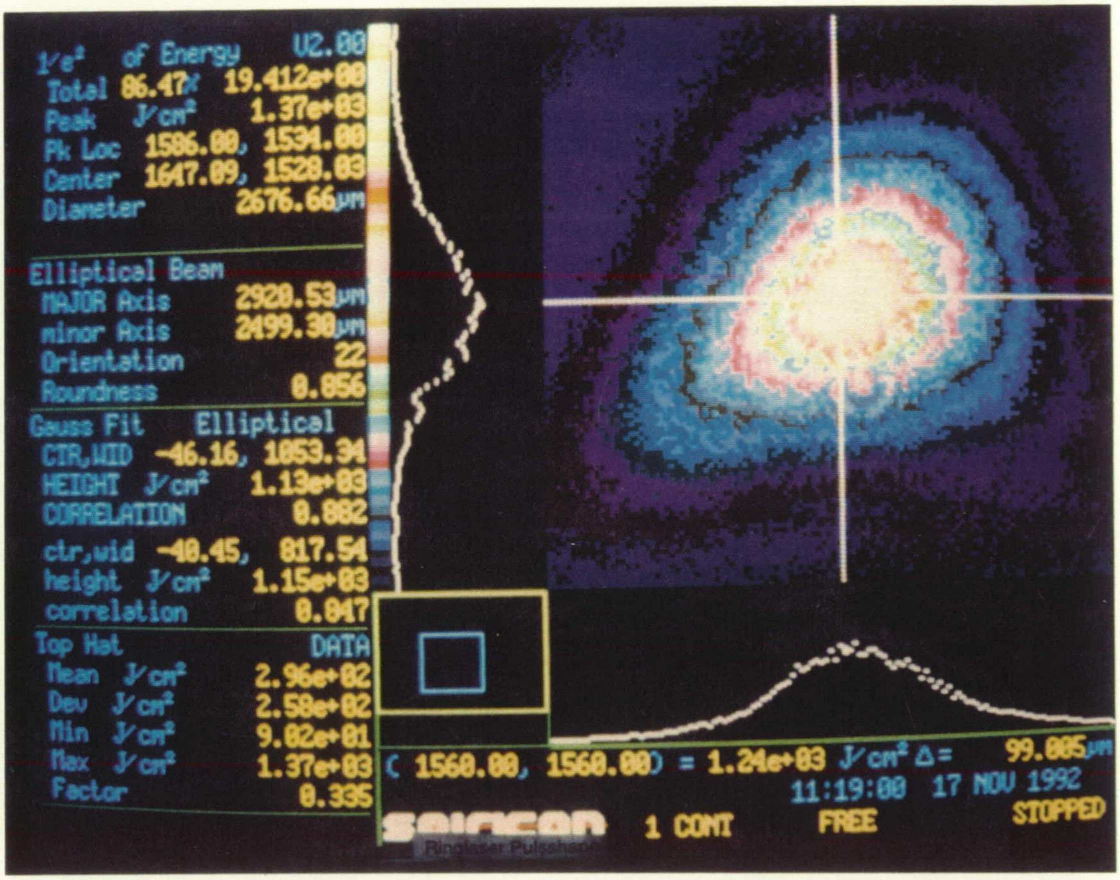

1/e² of Energy V2.00
Total 86.47% 19.412e+00
Peak J/cm² 1.37e+03
Pk Loc 1586.00, 1534.00
Center 1647.09, 1528.03
Diameter 2676.66µm
Elliptical Beam
MAJOR Axis 2920.53µm
minor Axis 2499.30µm
Orientation 22
Roundness 0.856
Gauss Fit Elliptical
CTR,WID -46.16, 1053.34
HEIGHT J/cm² 1.13e+03
CORRELATION 0.882
ctr,wid -40.45, 817.54
height J/cm² 1.15e+03
correlation 0.847
Top Hat DATA
Mean J/cm² 2.96e+02
Dev J/cm² 2.58e+02
Min J/cm² 9.02e+01
Max J/cm² 1.37e+03
Factor 0.335
(1560.00, 1560.00) = 1.24e+03 J/cm² Δ= 99.005µm
11:19:00 17 NOV 1992
1 CONT
FREE
STOPPED

FIGURE 14

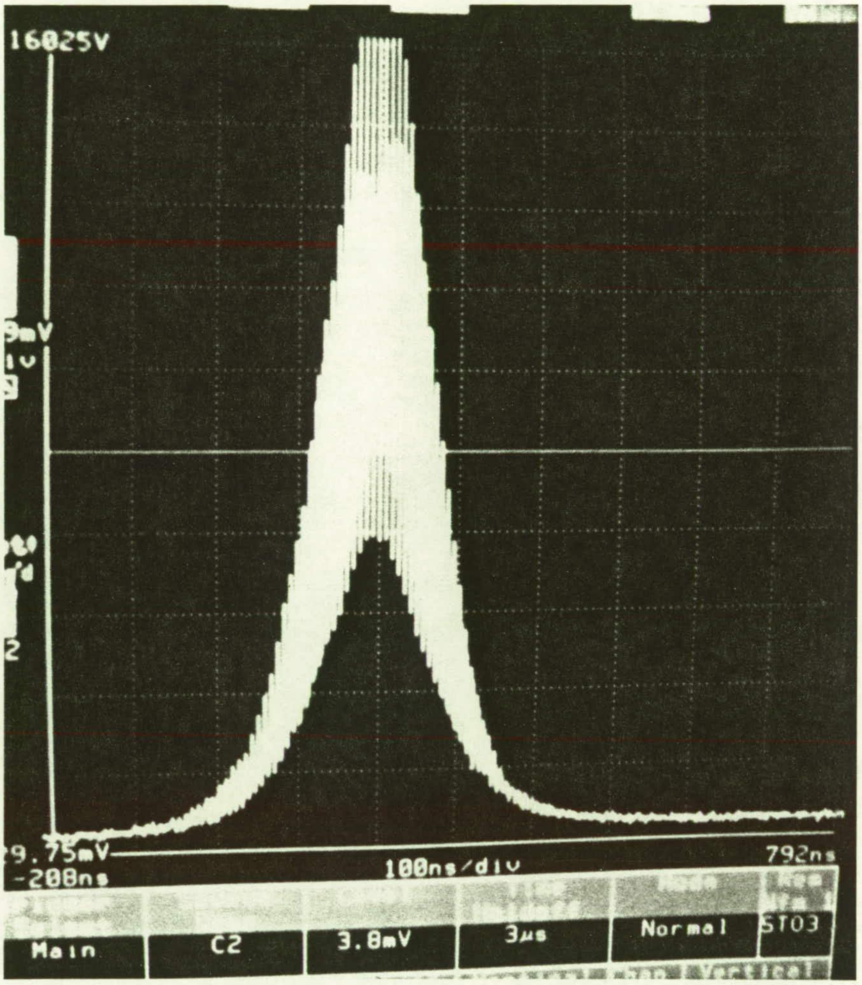
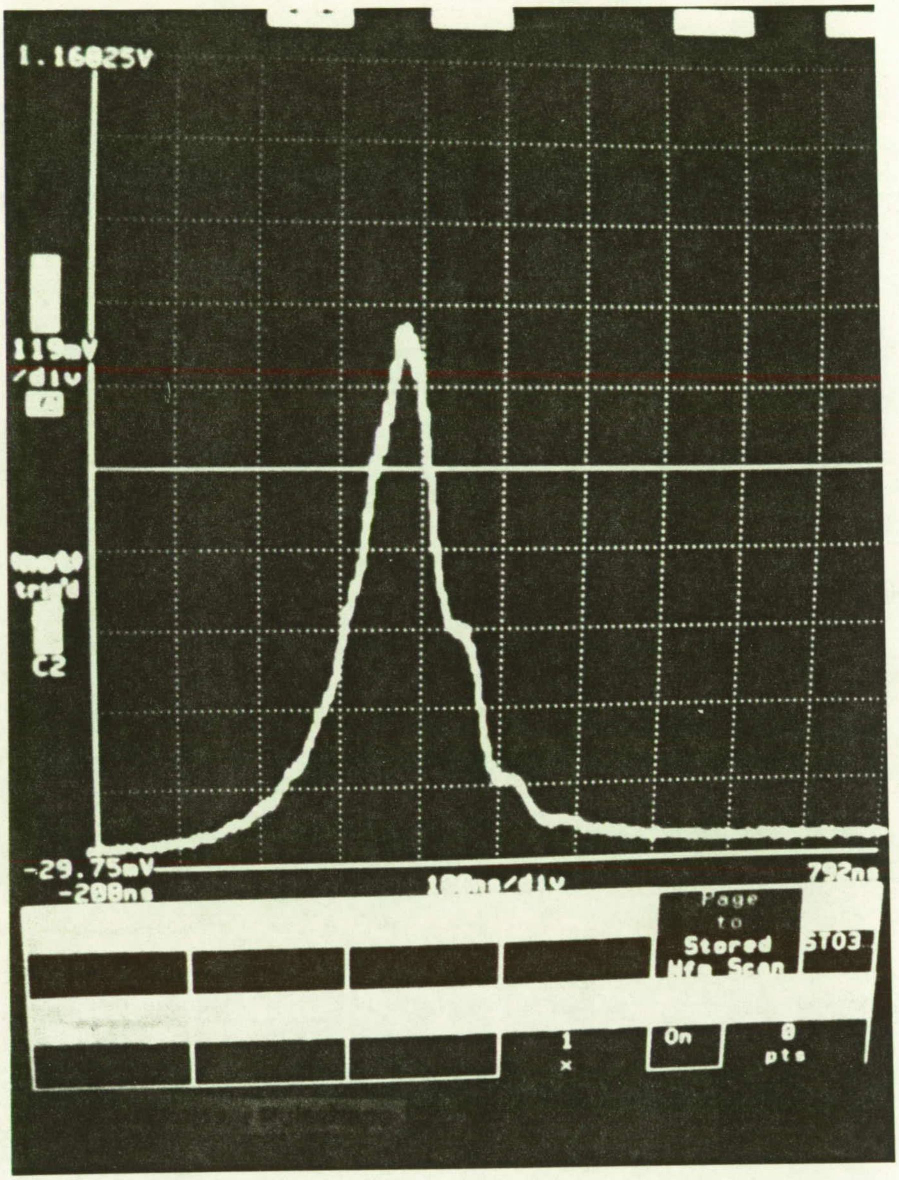

16025V
29.75mV
-208ns
100ns/div
792ns
Main
C2
3.8mV
3µs
Normal
ST03

FIGURE 15



1.16025V
119mV
/div
trig'd
C2
-29.75mV
-200ns
100ns/div
792ns
Page
to
Stored
Wfm Scan
ST03
1
x
On
0
pts

FIGURE 16

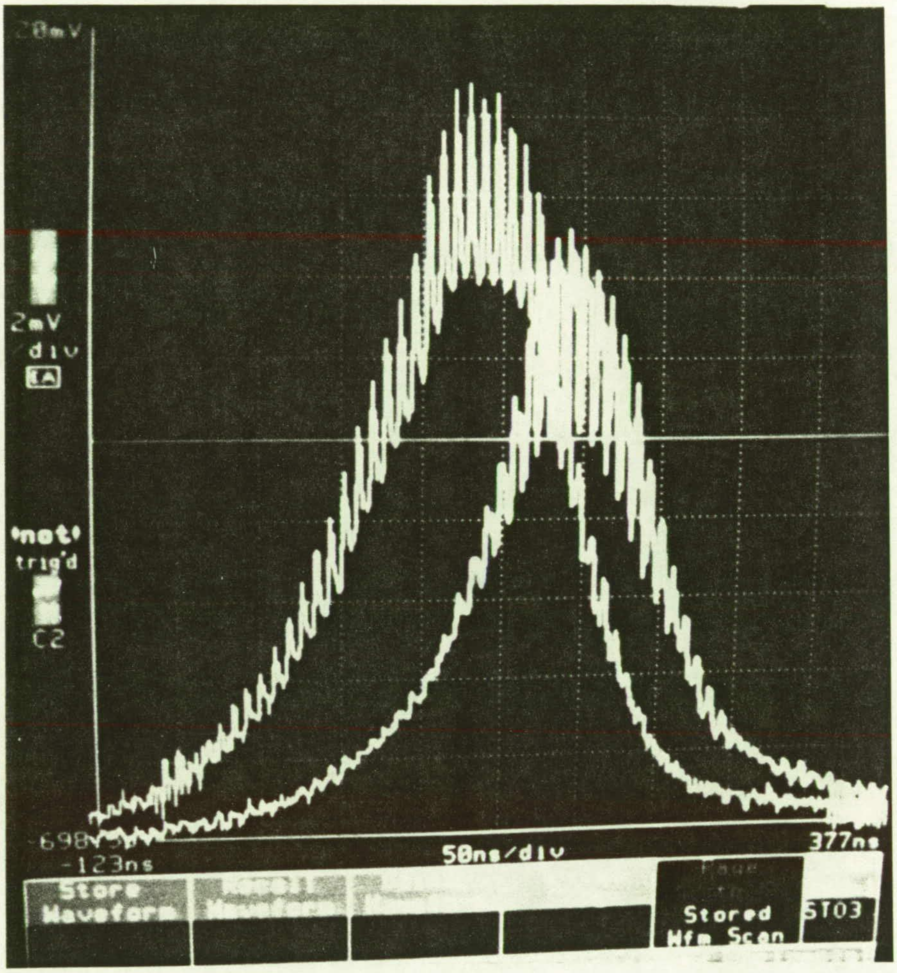

20mV
2mV
/div
'not'
trig'd
C2
-123ns
50ns/div
377ns
Store
Waveform
Stored
Wfm Scan
ST03

FIGURE 17

output of a 1 MHz-wide dye laser, to obtain linewidths of less than 20 MHz, where the transform limit would have been of order 5 MHz width. Second, Bischel and colleagues have demonstrated consistent collision-narrowing of the stimulated-Raman-scattered linewidth in hydrogen, obtaining linewidths as small as 100 MHz in some cases [W.Bischel *et al.* Phys. Rev. **A33**, 3113-3123 (1986)]. Third, MacPherson and colleagues, using Nd:YAG, have studied quantum fluctuations in the stimulated Raman scattering linewidth, and have demonstrated the statistical occurence of truly transform-limited pulses that, when accumulated over time, present an apparent average linewidth equal to the collision-narrowed width [D.C.MacPherson *et al.*, Phys. Rev. Lett. **61**, 66-69 (1988)]. The technique for reproducibly generating transform-limited pulses in systems such as we have described above will have to include, at least, such good mode control of the ring laser that, when the high gain Raman process starts, it starts with only those photons (or even just one photon) having the desired frequency. For 30-100 nsec pulse duration, the transform-limited linewidths are in the range 5-15 MHz, or (1.6 - 5.0) $\times 10^{-4}$ cm^{-1}. The ability to operate reliably at this level of pulse linewidth would benefit lidar remote sensing and other applications. Another level of resolution may be opened up by the application of solid state laser pulse-stretching methods to provide pulse lengths in the μsec range.

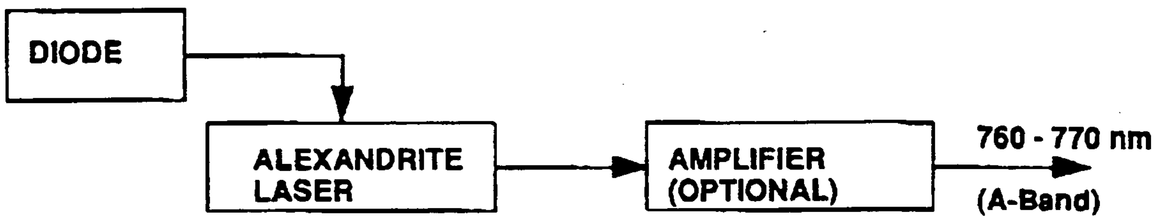
The advantages of stimulated Raman scattering as a central component of the systems we have studied are: high gain, high single-pass energy conversion (30-40% for first Stokes), even higher intracavity conversion (it can be as high as 70%, or 100% on a photon-for-photon basis), freedom from damage in the Raman medium (except for possible plasma breakdown at high power density), and the development of Light Age's gas- recirculating Raman cell that permits working with a wide variety of gases and Raman-shifted frequencies.

Fortunately, the alexandrite laser can be pumped directly, either with flashlamps or high power diodes, and does not require another relatively inefficient laser for this purpose. The marriage of such a tunable, solid state laser with diode laser injection seeding and nonlinear conversion processes can provide narrow band, high intensity light at nearly any wavelength. Applications of such systems to atmospheric lidar are shown schematically in **Figures 18** and **19**, for the oxygen "A-band" region, three band systems of the water molecule, and the sodium D-line region. A study by C.Gardner (Univ.of Illinois, private communication) shows that such alexandrite-based systems can, with considerable optical power, cover the wavelength range 370-770 nm for mesospheric lidar studies of the atomic species Fe, Mg, Ca^+, Al, Ca, Mg^+, Na, Li, and K.

For application to the study of atmospheric water vapor, it is clear that such alexandrite laser systems offer a very interesting option for DIAL measurements of H_2O in the tropical and midlatitude troposphere (720 nm band), that can then be Raman-converted to the 940 and 1140 bands for measurements of the lower concentrations of H_2O in the polar regions and the stratosphere. We have shown that a sound technical basis exists for further development of this capability, and specifically that intense radiation in the 940 band is generated with suitably narrow linewidth for water vapor measurement, even at the low temperature and pressure of the stratosphere.

Diode-Injected AlexandriteLaser: Lidar Examples

O_2 - DIAL ($\Delta\tilde{\nu}$ goal: 0.001 - 0.003 cm^{-1})



H_2O - DIAL ($\Delta\tilde{\nu}$ goal: 0.003 - 0.01 cm^{-1})

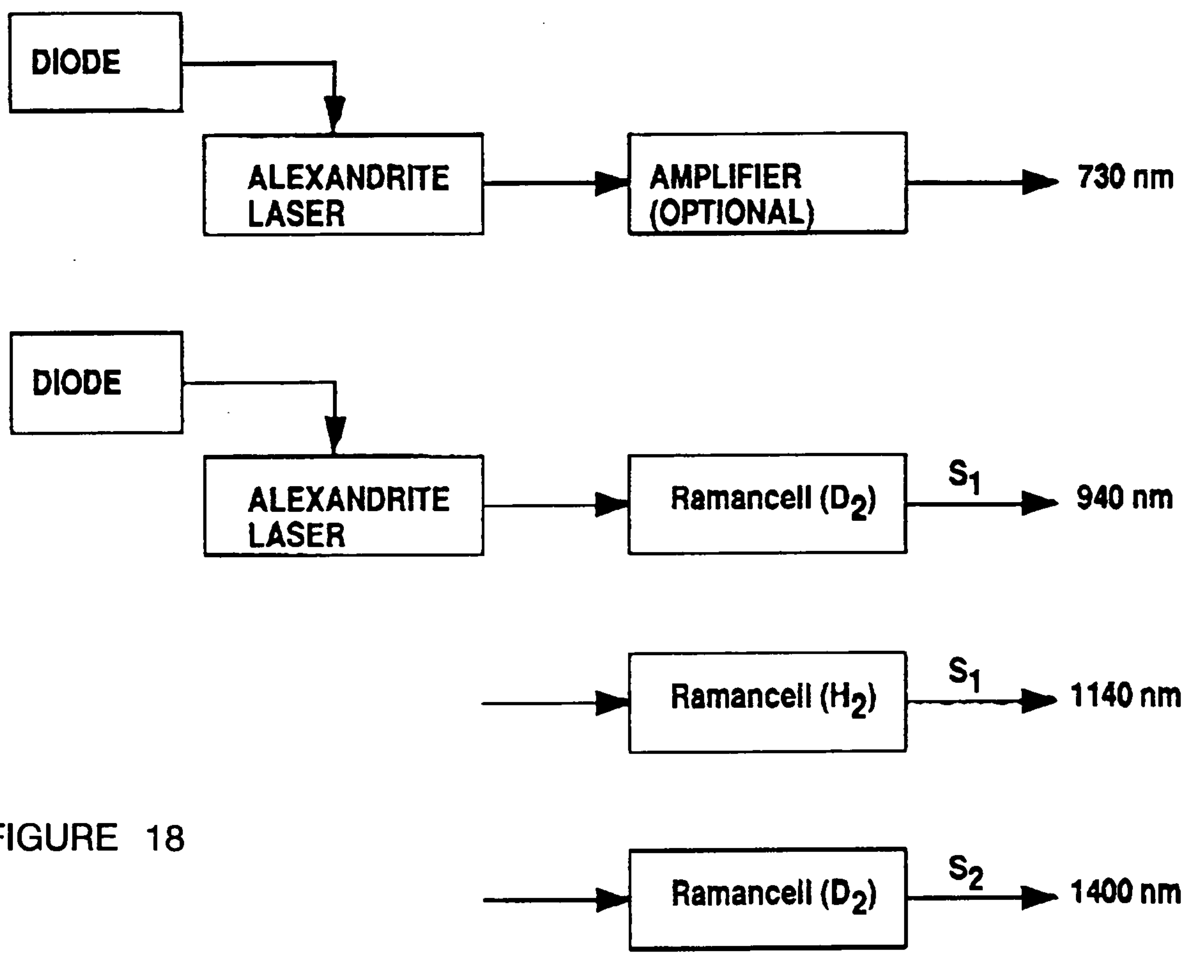


FIGURE 18

Na (D_2) ($\Delta\tilde{\nu}$ goal: 0.003 - 0.1 cm^{-1})

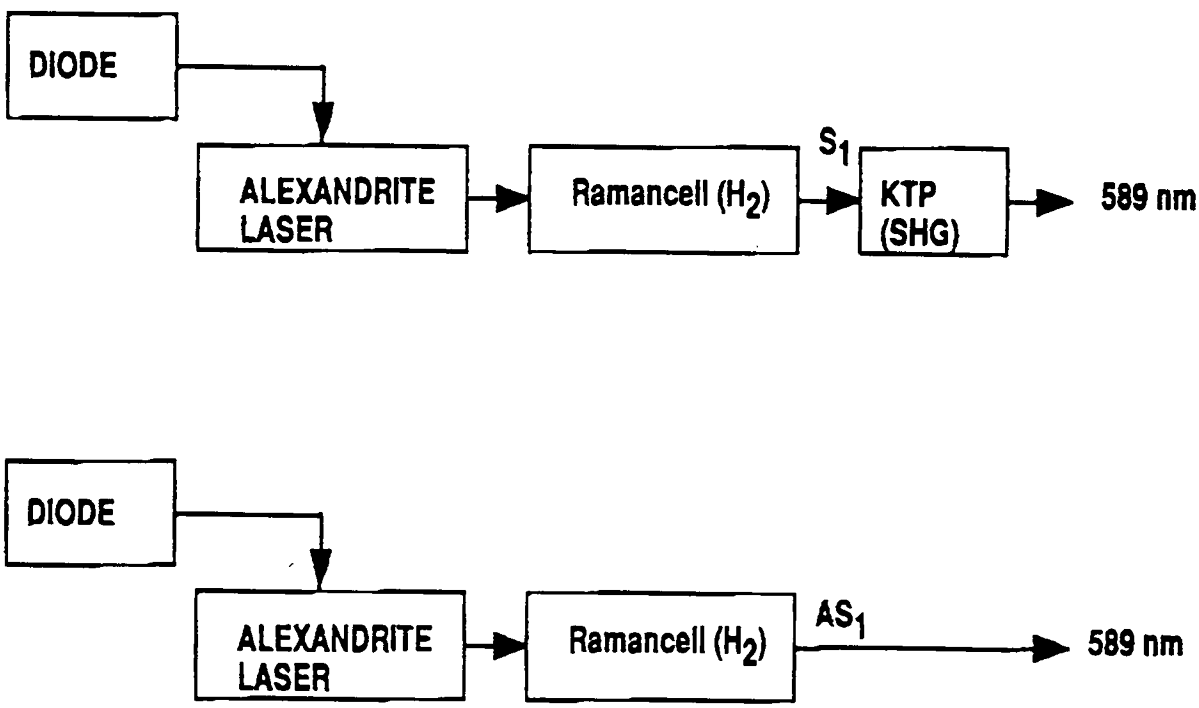


FIGURE 19

Appendix A

ABSTRACT FOR OSA CONFERENCE ON OPTICAL REMOTE SENSING OF THE ATMOSPHERE

Diode Laser Injection Seeded, Raman Shifted Alexandrite Laser Tunable Narrowband Lidar Source. St. Schmitz, and U. von Zahn, Univ. of Bonn, T. Wilkerson, Univ. of Maryland, D. F. Heller, and J. C. Walling, Light Age, Inc., 2 Riverview Drive, Somerset, NJ 08873; (908) 563-0600--- Diode laser injection seeded alexandrite lasers have been developed and their fundamental and Raman shifted output characterized. Application to water vapor, sodium, and other Lidars is discribed.

Diode Laser Injection Seeded, Raman Shifted Alexandrite Laser Tunable Narrowband Lidar Source. St. Schmitz, and U. von Zahn, Univ. of Bonn, T. Wilkerson, Univ. of Maryland, D. F. Heller, and J. C. Walling, Light Age, Inc..---

SUMMARY

The immediate purpose of this work is to develop robust, reliable, spectrally narrow light sources for optical remote sensing and specifically for certain atmospheric Lidar studies. One application is the study of the mesospheric sodium layer (80-100 km altitude) to determine altitude profiles of atomic density and temperature. For this purpose we have undertaken comparison of the Na (D_2) radiation at 589.159 nm produced by either anti-Stokes or frequency doubled first Stokes conversion of alexandrite laser wavelengths. A second application is the profiling of atmospheric water vapor by means of differential absorption Lidar (DIAL) utilizing alexandrite fundamental and Stokes shifted outputs in the 730 nm, 940 nm and 1140 nm bands of water vapor. The 1.4 μm water vapor band also is accessible using output from the Second Stokes shift of alexandrite (at 760 nm) radiation in D_2 gas.

In these and many other lidar applications, minimizing the linewidth of the lidar source is of paramount importance. Typically linewidths for these applications need to be in the 100-500 MHz range. For these applications adequate suitable narrowband laser sources have not previously been available. Here we describe the technical development of one very promising laser source. Concomitant with the source development certain fundamental issues regarding the physics of narrowband light generation with broadly tunable laser materials have been addressed as have concerns regarding limits to the faithfulness of narrowband Raman conversion processes.

Ring and standing-wave diode laser injection seeded alexandrite lasers have been Raman shifted in hydrogen, deuterium, and other gases and their outputs characterized. The fundamental output consists of single axial mode or few axial mode contributions depending on configuration. Most exciting is the ability of these laser sources to provide high average power, extremely narrowband light at most wavelengths in the UV, visible, and IR spectral regions. These sources appear to be extremely well-suited for most

atmospheric Lidar applications especially those requiring some degree of ruggedness and reliability.

Preliminary studies of the spectral output of the laser source were conducted at wavelengths near 780 nm using various commercially available, internal cavity diode lasers. The diode lasers produced between 5 and 100 mW of single frequency output and were continuously tunable, both by temperature and current, over ranges of a few to a few tens of GHz more. One particular (multi-quantum well design) diode has a continuous tuning range of approximately 150 GHz. Typical diodes have linewidths measured to be below 50 MHz. The output of the diode injection seeded alexandrite laser typically consists of between one and 4 longitudinal modes, depending on cavity configuration and seeding conditions. The single mode linewidth was measured to be below 80 MHz, limited by the resolution of our equipment. [The transform limited linewidth for pulses ranging from 30-100 ns in duration is 5-15 MHz.] The longitudinal mode spacing for both ring and standing-wave cavities is about 200 MHz. In these experiments no attempt was made to synchronize the laser cavity length to the diode frequency. Consequently the output typically consisted of contributions from a few cavity modes - dominantly one or two. High frequency mode beating was observed as modulation on the Q-switched pulse waveform and provided a signature for injection seeding. The sporadic single frequency pulses had the expected smooth pulse envelope. More that 200 mJ of pulse energy was obtained using multi-mode resonators and more than 30 mJ was obtained in a single transverse (TEMoo), single longitudinal mode beam. Pulse repetition frequencies were varied up to 25 Hz.

The output of the Q-switched, injection seeded alexandrite laser was Raman shifted in hydrogen and deuterium gases. The dominant characterization was done using hydrogen. Commercial Raman cells of a novel design (Light Age, Inc. 101 PAL/RC) were used. The cells of could be varied in length up to 1 meter and had integral focusing lenses that were nominally half the cell length. Cell pressure was varied but not necessarily optimized during these studies. The Raman shifted output consisted of several output (Stokes and anti-Stokes) beams at frequencies differing by 4155 cm-1, the Q(1) vibrational spacing in hydrogen. These beams were all collinear and tuned with the alexandrite pump laser frequency. Up to 5 orders of anti-Stokes beams and 2 orders of Stokes

beams could be observed by using hand held fluorescent cards and/or IR viewers. In many cases about 50% of the incident light was depleted in the conversion. More than 30% conversion into the first Stokes could be achieved. Conversion into the first anti-Stokes was more modest, but exceeded 5%.

The spectral output of the Stokes and anti-Stokes beams was extremely narrow, below our resolution limit. Our Fabry-Perot could easily resolve adjacent cavity modes of the alexandrite laser (200 MHz) but was limited in the determination of spectral bandwidth to about 350 MHz. This limit was about a factor of 4 smaller than the (spontaneous) Raman linewidth of hydrogen gas a the pressures used and demonstrates that substantial gain narrowing occurs in the conversion process. We expect but cannot yet resolve the transform limited Raman shifted pulses that may occur from time to time as found by MacPherson et al using fixed frequency laser pumps. These experiments demonstrate that diode injection seeded alexandrite lasers are robust and convenient sources useful for a great number of Lidar applications. Work is progressing to obtain linewidths below 100 MHz at 589 nm for atmospheric sodium studies of mesospheric winds and for operation in the 725-735 nm and 940 nm regions for water vapor Lidars.

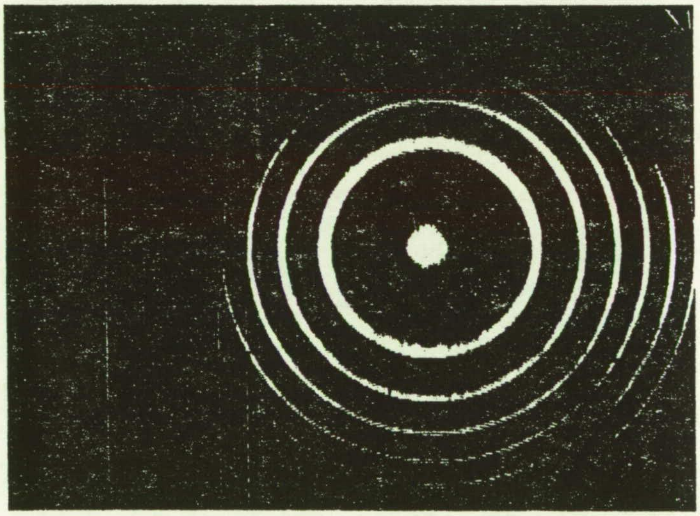


Figure 1: Fabry-Perot pattern from frequency doubled first stokes output
Free Spectral Range = 5 GHz.

Appendix B

ABSTRACT FOR CONFERENCE ON LASERS AND ELECTROOPTICS (CLEO'93)

Diode Laser Injection Seeded, Raman Shifted Alexandrite Laser Tunable Narrowband Lidar Source. D. F. Heller, and J. C. Walling, Light Age, Inc., 2 Riverview Drive, Somerset, NJ 08873; (908) 563-0600, T. Wilkerson, Institute for Physical Science and Technology, Univ. of Maryland, St. Schmitz, and U. von Zahn, Physikalisches Institut, Univ. of Bonn ---

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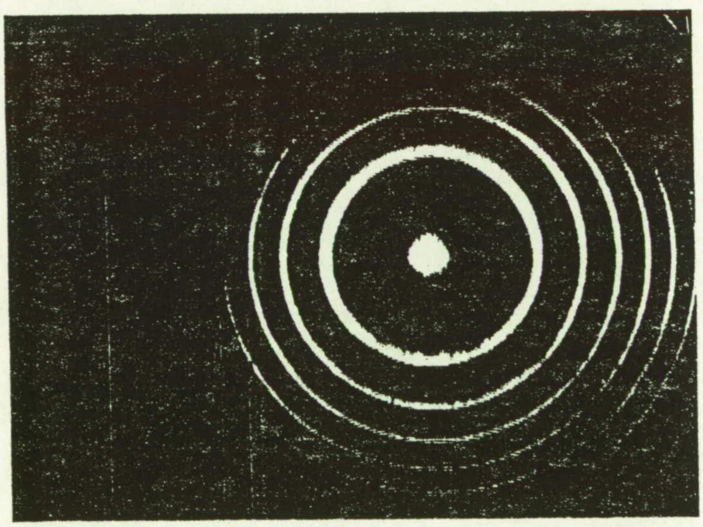

Figure 1: Fabry-Perot pattern from frequency doubled first Stokes output in hydrogen gas. Free Spectral Range = 5 GHz.